

THE ASSESSMENT OF TRACE ELEMENT CONTAMINATION OF THE
DRAINAGES FROM COAL CLEANING WASTES

by

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ABSTRACT

The mineral wastes produced by coal cleaning contain an abundance of toxic or potentially harmful trace elements. Although it has been well established that many of these toxic elements are present in the drainages from coal waste dumps, little is known about the quantities released into the environment from this source. The objectives of an ongoing research program at the Los Alamos Scientific Laboratory are to assess the nature and magnitude of the trace elements in the effluents from coal refuse materials, and to develop appropriate environmental control technologies as needed. ~~In this paper we present~~ recent experimental results ~~from this program~~ concerning the chemistry and aqueous leaching behavior of the trace elements in high sulfur coal cleaning wastes. ^{are presented} The emphasis ~~of the presentation~~ is on the interpretation of the experimental data that has led to the identification of the trace elements in coal refuse effluents that are consistently present in unacceptable quantities, and the development of environmental control strategies designed to abate or mitigate the problems of environmental concern that have resulted from these refuse drainages.

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INTRODUCTION

The mineral wastes from coal preparation and mine development constitute a major environmental problem. More than 3 billion metric tons of these materials have accumulated in the U. S., these are in coal waste dumps estimated to be between 3000 and 5000 in number of which half pose some type of health, environmental, or safety problem. Structural weaknesses in coal refuse banks have led to tragic landslides such as those at Buffalo Creek, West Virginia, and Aberfan, Wales, and the approximately 300 burning waste banks are a major source of air pollution. In addition to these problems, there is growing concern about environmental effects from the trace elements present in the highly mineralized acid drainage from coal refuse dumps that affect many thousands of miles of streams and waterways.

Although it has been established that the drainage from coal refuse dumps is often highly contaminated with trace or inorganic elements, little is known about the quantities of undesirable elements released into the environment from this source. Also development of the necessary control technologies for human and environmental protection requires quantitative evaluation of the extent and severity of the problem. As one of the steps in the solution to these problems LASL has been directed by the Department of Energy (DOE) and the Environmental Protection Agency (EPA) to assess the nature and magnitude of trace elements in the effluents from coal preparation wastes, and to identify the necessary control technologies.

The major activities in the LASL research program are,

- (1) assessment of the nature and magnitude of trace elements in the effluents from coals and coal preparation waste,
- (2) experimental identification of the chemistry of the trace constituents of environmental concern, and

- (3) identification and experimental verification of effective environmental control strategies to control the release of hazardous trace elements.

This paper describes the results of the research thus far carried out on coal wastes from the high sulfur coals of the Illinois Basin and some results of our studies on coal wastes from the Appalachian Region.

EXPERIMENTAL

1. Solid Waste Characterization

To understand why coals and coal preparation wastes release trace elements (i.e. all elements except C, H, S, N, O) in the amounts that they do, levels and mineral relationships of these elements have been studied in samples of coal preparation wastes from the Illinois Basin¹ and from the Appalachian Region.²

The Illinois Basin refuse samples were composed of clay minerals (largely illite and kaolinite), quartz, pyrite and marcasite. The average mineral content from three coal cleaning plants sampled is given in Table I. The Appalachian refuse that we used for our experiments differs in that it contains considerably less pyrite and marcasite, and a higher fraction of residual coal and unidentified matter. These data are also listed in Table I.

These studies of refuse structure and mineralogy revealed the great potential of these materials to release harmful quantities of trace elements. A large number of elements that are generally considered to be environmentally sensitive are present in these refuse materials in significant quantities ($> 30 \mu\text{g/g}$). Included among these are fluorine, aluminum, manganese, iron, cobalt, nickel, copper, zinc, arsenic, and lead. Although the relative amounts of some of these components are seemingly small, the absolute quantities that are available in a large or active waste dump might have grave consequences in the surrounding environment if they were to be released and concentrated by natural processes.

TABLE I
MAJOR MINERALS IN COAL PREPARATION WASTES[†]

<u>Mineral</u>	<u>Illinois Basin (Avg. for 3)</u>	<u>Appalachian Region</u>
Kaolinite, $(Al_2Si_2O_5(OH)_4)$	12	11
Illite, $(K Al_2(Si_3Al)O_{10}(OH)_2)$	14	19
Other clays	11	5
Pyrite, Marcasite, FeS_2	26	~ 1
Quartz, SiO_2	21	20
Calcite, $CaCO_3$	3	1
Gypsum, $CaSO_4 \cdot 2H_2O$	2	3
Other (coal, unidentified species)	11	40

[†] wt%

A list of trace elements and minerals with which these are associated, as revealed by our researchers with field samples from three Illinois Basin coal preparation plants is shown in Table II

2. Characterization of coal waste leachates

Static and dynamic leaching experiments have been performed to evaluate the trace element behavior of these coal wastes under simulated weathering conditions. These experiments were done to provide information needed to predict quantitatively the trace element levels in the drainage from coal refuse dumps or disposal areas and to identify those elements of environmental concern.³

One of the more important characteristics of the refuse materials is their ability to produce acidic leachates during aqueous leaching as a result of the oxidative degradation of the pyrite and marcasite minerals. This is true for leachates from the high sulfur Illinois Basin refuse (leachate pH, 2-4) and for leachates from the low sulfur Appalachian region refuse (leachate pH ~ 3).

TABLE II
SUMMARY OF TRACE ELEMENT - MINERAL ASSOCIATIONS IN REFUSE
FROM THREE ILLINOIS BASIN COAL PREPARATION PLANTS

<u>Element</u>	<u>Mineral Association</u>	<u>Element</u>	<u>Mineral Association</u>
H	coal	Co	clays
Li	clays	Ni	clays
Be	quartz	Cu	clays/pyrite/marcasite
B	clays	Zn	clays
C	coal/carbonates	Cd	clays
N	coal	As	pyrite/marcasite
V	phosphates/carbonates	Se	pyrite/marcasite
Ka	clays	Br	coal
Hg	carbonates/clays	Rb	clays
Al	clays	Y	phosphates/clays
Si	quartz/clays	Zr	oxides/clays
P	phosphates	Mo	pyrite/marcasite
S	pyrite/marcasite/gypsum	Cd	clays
Cl	coal/clays	Ce	clays
K	clays	Ba	clays/phosphates
Ca	carbonates/clays/gypsum/phosphates	Rare earths	phosphates/clays
Sc	clays	Hf	clays
Ti	clays/oxides	Ta	clays
V	clays/quartz	Hg	pyrite/marcasite
Cr	clays	Tl	pyrite/marcasite
Mn	clays/carbonates/pyrite/marcasite	Th	phosphates/clays
Fe	pyrite/marcasite/clays/carbonates	U	phosphates/clays

These acid leachates are very efficient in dissolving or degrading many of the minerals in the refuse, thus releasing the trace elements associated with them. Figure 1 shows the relation between pH and total dissolved solids as determined for some of the experimental leachates produced using coal wastes. It is clear from the figure that the highly acid solutions that result from pyrite oxidation can produce highly contaminated leachates.

Experiments have been performed to simulate the intermittent rainfall and weathering to which refuse dumps are subjected. These researches have revealed that this alternate oxidation and leaching of the pyrite in the refuse is a most effective way to generate acid leachates and indeed these intermittently leached coal waste piles pose a far greater pollution threat, in terms of both quantity and time, than those wastes that are always in contact with water or

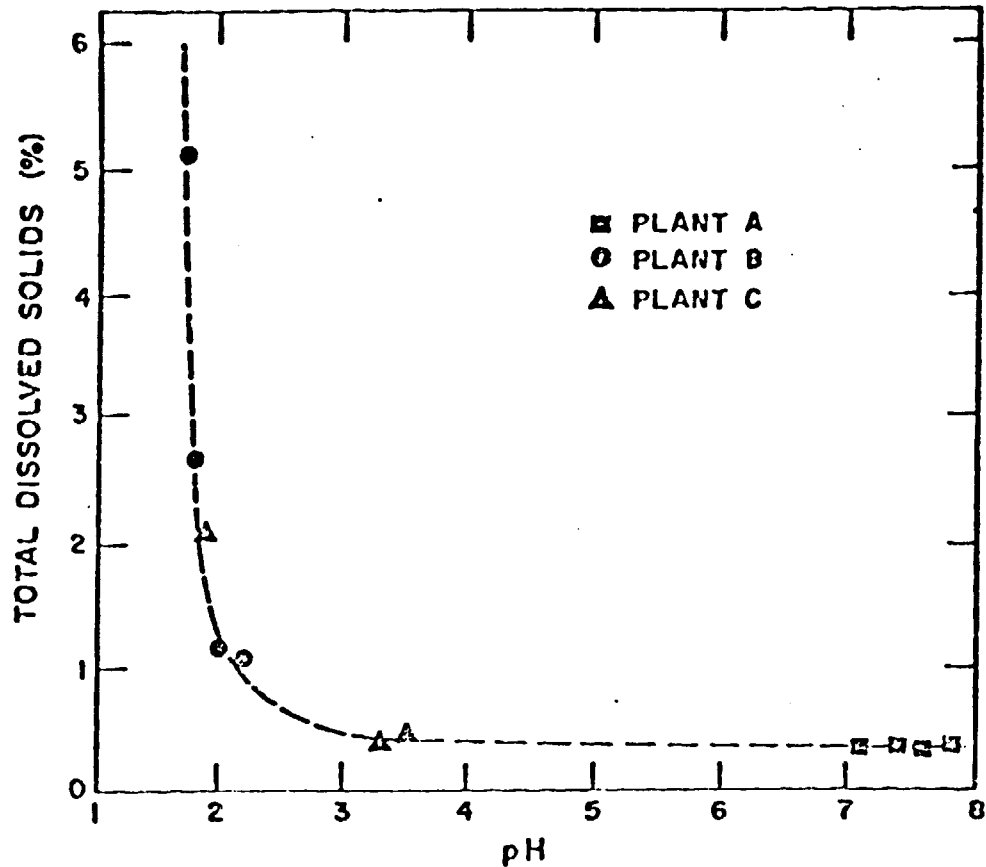


Fig. 1. The relationship between pH and total dissolved solids for leachates from static leaching experiments with Illinois Basin coal refuse.

or otherwise isolated from air. The experimental data on trace element leachability that we generated as a result of these simulated weathering and leaching studies have also been compared with similar data for actual refuse dump drainage from diverse points in the Illinois Basin. The high level of agreement between the two sets of results indicates convincingly that the laboratory leaching tests realistically simulate refuse dump conditions!

Our investigations have shown that aqueous leachates from the high sulfur coal refuse of the Illinois Basin contained nine elements (F, Al, Mn, Fe, Co, Ni, Cu, Zn, and Cd) in environmentally hazardous concentrations.⁴ The criteria used for identifying trace elements of environmental concern have been published by the U. S. Environmental Protection Agency⁵ in a report that defines

multimedia environmental goals (MEGs) and the minimum acute toxicity effluents (MATEs). These data are displayed along with other trace elements identified in this work in Fig. 2.⁶ Leachates derived from the low-sulfur Appalachian coal

Figure 2. Trace Elements of Environmental Concern in Coal Waste Drainages.

(H)																	He
(Li)	(Be)											(B)	C	N	O	(F)	Ne
(Na)	(Mg)											(Al)	(Si)	P	S	(Cl)	Ar
(K)	(Ca)	(Sc)	(Ti)	(V)	(Cr)	(Mn)	(Fe)	(Co)	(Ni)	(Cu)	(Zn)	(Ga)	(Ge)	(As)	(Se)	Br	Kr
(Rb)	(Sr)	(Y)	(Zr)	(Nb)	(Mo)	(Tc)	(Ru)	(Rh)	(Pd)	(Ag)	(Cd)	In	(Sn)	(Sb)	(Te)	I	Xe
(Cs)	(Ba)	(La)	(Hf)	(Ta)	(W)	(Re)	(Os)	(Ir)	(Pt)	(Au)	(Hg)	Tl	(Pb)	Bi	Po	At	Rn
Fr	Ra	Ac															

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 OK

(Ce)	Pr	Nd	Pm	(Sm)	(Eu)	Gd	Tb	(Dy)	Ho	Er	Tm	(Yb)	(Lu)
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(Th)	Pa	(U)	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr
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Shaded elements exceed MATE values
Circled elements were analyzed

refuse were also analyzed in terms of EPA's MEG/MATE system for classifying potentially hazardous contaminants.² These researches have identified Ni, Fe, Mn, Cu, Al, and Zn as the leachate pollutants of greatest environmental concern in the Appalachian region coal wastes that we have studied.

3. Environmental Control Technology Studies

It is necessary, but not sufficient to only assess the nature and magnitude of trace element contamination in coal waste drainages. A necessary adjunct to an experimental program of this nature is a concurrent laboratory evaluation and identification of environmental controls for abatement or diminution of the pollution in these drainages. These laboratory investigations have been carried out and the results classified into three environmental control strategies.

These strategies may be described as follows:

- o Treatment of the refuse (before disposal) to remove or immobilize mobile trace elements of environmental concern.
- o Treatment of the refuse (at the time of disposal) with neutralizing agents, absorbents or sealants to control the release of undesirable trace elements from the refuse pile.
- o Treatment of the contaminated drainage (after disposal) to reduce trace element concentrations to environmentally acceptable levels.

Clearly there are many possibilities to consider in each category. For this presentation we will describe the basic control technology, that we have identified in our researches, that holds the greatest promise in each of these areas.

Pre-disposal calcining to immobilize refuse constituents

The possibility that the release of toxic trace elements into the environment can be controlled by pre-treatment of coal preparation wastes has been

investigated experimentally. One approach examined is that of heat-treating the wastes to form what appears to be a glass-like structure that apparently immobilizes the trace elements in the refuse matrix and prevents their escape.

Several sets of calcining and leaching experiments were conducted to determine optimum heat treatment conditions necessary to chemically immobilize the potentially toxic trace elements in the refuse matrix. These experiments were performed using high sulfur coal preparation waste from a plant in the Illinois Basin. The waste was ground to -20 (0.84 mm opening) mesh and calcined in air at 873K, 1073K, 1273K, and 1473K for 2 h. The calcined residues were then statically leached (stirred with distilled H₂O) for a period of 48 h using a ratio of 4 \pm H₂O to 1 kg of calcined refuse, and pH's and total dissolved solids determined.

The effect of the refuse calcining treatments on the mineral composition of the refuse is illustrated by Tables III and IV. Table III delineates the changes in refuse mineralogy that have occurred as a function of heat treatment. Table IV presents the mineral conversion reactions that occur at high temperature. It is seen from Table III that the two most environmentally active species,

TABLE III
EFFECTS OF CALCINING ON COAL REFUSE MINERALOGY

TEMPERATURE	PYRITE	CLAYS	QUARTZ	CALCITE	HEMATITE	CORUNDUM
Uncalcined	Present	Present	Present	Present	-	-
873K	-	Present	Present	-	Present	-
1073K	-	Present	Present	-	Present	-
1273K	-	-	Present	-	Present	-
1473K	-	-	Present	-	Present	Present

TABLE IV
HIGH TEMPERATURE MINERAL TRANSFORMATIONS

Clays	Complex Aluminosilicates
Calcite	$\text{CaO} + \text{CO}_2 \uparrow$
Pyrite or Marcasite	$\text{Fe}_2\text{O}_3 + \text{SO}_2 \uparrow$
Quartz	Quartz

pyrite (marcasite) and calcite, have been transformed to high temperature phases by 873K. By 1273K even the clay minerals have been converted to structurally indefinable aluminosilicates. The x-ray diffraction analyses reported in Table III confirms that the mineralogical transformations listed in Table IV have been effected by heat treatment in the range of 1073 to 1273 K. Physically the samples calcined at 1273K were fused or sintered at particle surfaces.

The success of the calcining treatment at reducing trace element releases during refuse leaching is illustrated by the data in Table V, which lists trace element data of environmental concern from a comparison leaching test of calcined and uncalcined refuse samples. The refuse samples referred to in the table had been subjected to static leaching for 48 h. It is seen that the concentrations of the group of toxic elements listed are reduced in the leachates from the calcined refuse by as much as two orders of magnitude over the concentration in the raw refuse leachates. Leaching experiments were also conducted using acid solutions (pH < 2) with essentially the same results.

Calcining of acid coal refuse materials prior to disposal could produce several beneficial effects. Foremost among these is the conversion of an active, highly polluting waste material into a chemically and geologically inert mass that can be easily and safely disposed almost anywhere with ordinary

TABLE V
TRACE ELEMENT LEACHABILITY OF A HIGH SULFUR COAL
REFUSE SAMPLE CALCINED AT 1273K

<u>Element</u>	<u>Uncalcined Refuse</u>	<u>Calcined Refuse</u>
Al ^a	100	0.4
Fe	600	< 0.03
Mn	5.8	0.03
Co	2.8	< 0.01
Ni	4.8	0.01
Cu	0.10	0.01
Zn	2.8	0.05
Cd(ppb)	68	0.3
pH	2.9	8.0
TDS(%)	1.4	0.2

^aLeachate concentration reported as ppm unless otherwise noted (50 g crushed refuse/200 ml H₂O/48 h)

landfill practices. In so doing calcining presents a highly effective and probably permanent solution to a most difficult waste control problem. A corollary to this is that the calcined refuse materials might well be classified as nonhazardous under the criteria developed in conjunction with the Resource Conservation and Recovery Act (RCRA). This circumvents the need to meet the cumbersome and costly permit and performance requirements that RCRA dictates for the disposal of hazardous wastes, a category into which much, if not all, untreated high sulfur refuse materials will probably fall. Also, there is high potential for the recovery of by-products in connection with refuse calcining

that does not exist for many of the other control technology schemes. Potentially recoverable products include sulfur, iron, and aggregate materials.

Codisposal of wastes with alkaline materials

One of the major conclusions from our earlier studies of the environmental behavior of coal refuse materials concerned the importance of pH in controlling trace element releases during refuse leaching. In all instances when leachate pH was maintained at or near the neutral point, only minimal amounts of trace elements were solvated by the leachates. Conversely, when oxidative degradation of the pyritic materials in the refuse caused leachate acidities to build up, substantial quantities of such environmentally troublesome elements as Fe, Al, Mn, Cu, Co, and Ni were solvated by the acid leachates. This marked dependence of the degree of trace element contamination of refuse leachates on leachate pH suggested that a fruitful means of preventing trace element releases from discarded refuse materials would be to add neutralizing agents to the refuse at the time of disposal to reduce leachate acidity, in situ, as it is formed.

Column leaching experiments that utilize mixtures of powdered lime and high sulfur refuse were conducted to test the effectiveness of this in situ neutralization concept. These experiments proved to be very fruitful. For these experiments, powdered lime in varying amounts (3 to 50 g) was slurried in 150 ml of distilled water with -9.5 mm high-sulfur coal refuse (530 g, from Illinois Basin Plant B). The resultant mixture was subsequently dried in air at 323K and recrushed to -9.5 mm particles. Four different lime concentrations were employed, 0.5, 1.5, 3, and 10% (by weight).

Column leaching experiments were conducted with about 500 g of each of the above samples to determine the effects of the lime additions. The refuse mixtures were packed into pyrex columns 0.4 m long by 0.05 m diameter and subsequently leached with distilled water at a flow rate of .008 ml/s until

more than 4 ft water had been passed through the refuse beds.

A consistent pattern of the effects of the lime additions emerges from these data. The addition of 0.5 and 1.5 wt% lime to the acid refuse had only a small influence on leachate pH and trace element concentration because the acid neutralization provided by these amounts of lime was overwhelmed by the acid generating capability of the refuse. The additions of 3 and 10 wt% of lime, on the other hand, did indeed effectively counteract the acid properties of the refuse; the pH of the leachates for these two systems is higher, TDS values are relatively low, and the trace element concentrations are depressed.

The system containing 3 wt% lime is especially interesting because a leachate pH of 7 was maintained for nearly the entire duration of the continuous part of the leaching experiment (until 4.2 ft had been passed through the column). TDS values for this refuse-lime combination were also very respectable (ranging downward from about 0.6 wt%) especially considering that the dissolution of the lime itself adds substantially to the dissolved solids content of the solution. By the end of the leaching experiment, concentrations of troublesome trace elements, especially Fe and Mn, had been reduced to environmentally acceptable levels.

The codisposal of alkaline agents such as lime with acidic coal refuse materials does appear to be an attractive option for controlling trace element contamination of disposal area drainages. The technique is only moderately costly (our 1978 estimate was \$0.50 to \$1.00 per metric ton of cleaned coal) and appears to be a highly effective means of preventing the release of a contaminated drainage from coal refuse dumps. The technology for mixing alkaline agents with coal refuse materials should be relatively simple and is immediately effective.

There are however a few uncertainties associated with the use of alkaline additives for coal refuse materials. One uncertainty involves the long term effectiveness or permanency of the method. The durability and immobility of the alkaline additives over long geologic periods must be demonstrated. Another question associated with codisposing alkaline additives and high sulfur coal refuse materials concerns the RCRA classification of the resulting refuse-additive mixtures. It is not clear at this time whether such a mixture would be classified as hazardous or nonhazardous. As pointed out earlier, a hazardous RCRA designation could be quite costly for the disposal site operator. Another somewhat negative aspect of refuse codisposal with alkaline agents as compared to refuse calcining, for example, is its low potential for by-product recovery. The lack of such potential, of course, negates the possibility of offsetting environmental costs with recovered product value.

Post-disposal control of trace element contamination of refuse dump drainage

Water treatment is the best known and widely practiced of control technologies. While we have evaluated known and recognized control methods of water treatment for the coal waste drainages, (ion exchange, reverse osmosis, oxidation, chelation), we are most interested in controls that are realistic in terms of cost and ease of implementation; these are most necessary considerations in view of the enormous generation rate (greater than 100 million metric tons per year) of the coal wastes. One area that we have concentrated on is pH control by means of alkaline neutralization. The effect of pH on drainages from high sulfur coal wastes in the Illinois Basin has been investigated extensively during the progress of our investigations, and in Table VI we show some values of TDS vs pH derived from experiments conducted on our laboratory generated leachates. During these studies, coal wastes from three different coal cleaning plants were used and the table represents overall averages.

TABLE VI
EFFECT OF pH ON TOTAL DISSOLVED SOLIDS
FOR HIGH SULFUR COAL CLEANING WASTES

pH	TDS (%)
1.7	5.1
2	1.2
3	0.4
5	0.3
7	0.3

Alkaline neutralization is currently used extensively to treat acid drainage from coal mines. While it is well known that alkaline neutralization is very effective in controlling the acid and overall salt compositions of mine waste waters, the degree of control that this method exerts over some of the more highly leachable toxic trace elements remains to be established. Elaboration of this latter point is the basis for the study that we conducted in this area.

The experiments that we conducted were basically titrations in which limestone, lime, or lye (NaOH) were added to one liter of contaminated refuse drainage until a predetermined pH value was reached. The solutions (or slurries) were allowed to sit overnight, were filtered, the pH's were measured, and the compositions of the resulting solutions were analyzed.

As are the other control techniques discussed in this report, alkaline neutralization has been shown to be an effective method for reducing or abating trace element contamination of coal refuse drainage. The projected costs for such a treatment are relatively low (\$0.10 to \$0.80 per metric ton of cleaned coal) and the technique is relatively easy to apply, as evidenced by the large number of neutralization plants already in operation to treat acid mine drainage.

However, in spite of the low cost and ease of application, alkaline neutralization has some rather considerable disadvantages. For example, its use never really treats the source of contamination (i.e., the refuse itself) and treatment of drainage from the disposal site may be needed almost indefinitely. Also, although the standard refuse disposal practice at present involves burying the refuse on top of impermeable liners, such as clay, to channel refuse dump drainage into treatment areas, it is very difficult to ensure that polluted drainage will not eventually escape through or around these liners and thus negate the effectiveness of this method. Another consideration that may make alkaline neutralization less attractive involves the cost associated with meeting RCRA requirements. There is a possibility that waste materials disposed of in a way which produced contaminated drainage will be classified as hazardous. Thus the apparent low cost of alkaline neutralization may have to be tempered with additional costs needed to meet RCRA permit and performance requirements. Finally there is little opportunity for by-product recovery during or subsequent to neutralization treatment. Thus the potential for realizing economic gain in this way is low.

SUMMARY

The results of these researches make clear that coal preparation refuse drainages from the high sulfur wastes of the Illinois Basin and wastes with low sulfur contents from the Appalachian Region release acid and trace elements into surface and ground waters in concentrations that are environmentally unacceptable. Laboratory analysis of coal wastes and coal waste leachates have yielded information on acid formation, release mechanisms, and trace element concentrations in sufficient detail to allow us to address key environmental control issues in a controlled and methodical manner. These considerations have resulted in a control scenario in three parts; waste

treatment before disposal, waste treatment during disposal, and effluent treatment after disposal. An example of each method has been discussed along with the advantages, disadvantages, and tradeoffs that must be taken into account in any realistic environmental control evaluation. These collected conclusions are displayed in the following matrix summary (Table VII).

TABLE VII
OVERVIEW OF THREE ENVIRONMENTAL CONTROL
OPTIONS FOR COAL PREPARATION WASTES

	<u>Calcining</u>	<u>Lime Codisposal</u>	<u>Alkaline Treatment</u>
Cost	High	Low	Low
Effectiveness	Excellent	Good	Good
Complexity	High	Moderate	Moderate
By-Product Potential	High	Low	Low
Treatment duration	Short	Short	Long
Permanency	Good	Uncertain	Poor
RCRA potential	Non-hazardous	Uncertain	Hazardous

Future work in this program will attempt to expand our knowledge of coal wastes and their drainages in general, and to perfect more realistic environmental simulation experiments both in terms of conditions and of scale-up. These will lead to identification and evaluation of ever more realistic environmental control technologies.

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